THERMAL NEUTRON POWER REACTOR COOLANTS

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by

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PREFACE

Man's progress toward a higher civilization and a better way of life
is correlated closely with the energy available for his use. The discovery
of each new energy source has brought about a revolutionary advancement in
scientific and technical progress spurred on by man's insatiable desire to
make use of mechanical energy and lessen his physical burdens. Since the
turn of the twentieth century a new force has become available. Nuclear
fission, discovered in 1939 and controlled in 1941, has opened the door to
virtually an infinite source of energy. The struggle now is to make this
new source of energy available for useful work.

Current developments in nuclear reactor technology indicate that power
from the atom will soon be available. Reactors can be built to release
nuclear energy at controlled rates. The problem remaining is to remove
the energy released with the maximum efficiency attainable. It is hoped
that this paper will indicate the magnitude of this problem and some ave-
nues toward its solution.

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CHAPTER I

INTRODUCTION

1. Problem stated.

Simply stated, the problem of converting the energy released by the fissioning nuclei in a chain reacting pile to a useful form for work or power is a problem of thermodynamics. The only known method of making the conversion is through a heat cycle in which the fission energy is absorbed as heat in a reactor, transferred to a coolant in the reactor and transported by the coolant out of the reactor, where it is available for use in a heat engine to generate power in conventional forms. The efficiency of conversion is given by the thermodynamic relationship,

$$E = f \; \frac{T_{in} - T_{out}}{T_{in}} \times 100$$

where E is the conversion efficiency, f is the fraction of Carnot efficiency attained by an actual heat engine and T is the absolute temperature at which heat is supplied to (T_{in}), or exhausted from (T_{out}), the system. Since the fission chain reaction can be made to go at extremely high temperatures, it would appear that exceedingly efficient recovery of nuclear power is possible. Unfortunately this is not true, as metallurgical and engineering considerations impose a practical limit on the temperature at which heat can be extracted from a reactor. The problems of nuclear power recovery are then those usually associated with operations at high temperatures and high heat removal rates, compounded by requirements for considering the nuclear properties of structural materials and coolants, and by the intense radiation associated with the fissioning chain reaction.

In the nuclear power cycle the reactor in a sense replaces the boiler fire box of the conventional steam plant, for it is here that the nuclear

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fuel is burned and converted into heat. The coolant corresponds to the hot gases of combustion, which transport the heat to the boiler tubes. The whole problem of the design of reactors for power generation then revolves about the selection of a coolant for the most effective heat removal at the highest temperatures permitted by available structural materials.

Of the several reactor types which hold promise for developing nuclear power, the majority operate on thermal neutrons, and it is in this field of thermal reactors that the greatest amount of experience in reactor development is available. While it would be manifestly impossible in a paper of reasonable length to discuss the design features of the various thermal reactors having power production potentials, it is felt that some value can be derived from a qualitative discussion of the properties of materials available for use as thermal reactor coolants.

CHAPTER II

THERMAL REACTOR COOLANTS

1. Ideal coolant properties.

The requirements for a coolant in a thermal reactor designed for power
production are somewhat stringent. Not only must the material selected
have good thermal properties and be non-corrosive at working temperatures,
but it must also have desirable nuclear properties, and be stable to nuclear
radiation. The ideal coolant would be one that is non-corrosive and chemi-
cally inert and has in addition all of the following properties:

    a. Low melting and high boiling points.

    b. Large heat transfer coefficient.

    c. High density and specific heat.

    d. Low neutron capture cross section.

    e. High moderating power.

    f. Radiation stability.

    g. Low level of induced radioactivity.

Unfortunately the ideal coolant does not exist, but the selection of a
coolant for a given reactor is facilitated by an understanding of the rela-
tive merit of the above properties.

Non-Corrosiveness and Chemical Inertness: Corrosion is a more serious prob-
lem in reactor coolants than in conventional heat systems. Because of the
high residual radioactivity in a reactor core after shutdown the problem of
maintenance or repair is tedious and expensive. Reactor components, par-
ticularly the cooling system, must therefore have a long life expectancy,
and corrosion must be minimized. Furthermore the presence of corrosion pro-
ducts in the coolant will adversely affect neutron economy within the core,

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and magnify the problem of shielding the cooling system external to the reactor.

A chemically inert coolant would be relatively non-corrosive. It would, in addition, eliminate the possibility which exists in the use of certain liquid metal coolants of a chemical explosion on mixing of coolant and the working fluid of the steam plant.

Melting and Boiling Points: A low melting point, preferably below room temperature, facilitates coolant handling during periods of shutdown for maintenance or repair; it minimizes or eliminates the necessity for preheater equipment in the cooling system; it reduces the possibility of a freezing casualty in any part of the cooling system, and it insures the attainment of the low optimum coolant inlet temperature.

A high boiling point permits attainment of increased thermal efficiencies without resort to high pressure systems. The boiling temperature of the coolant at atmospheric pressure should equal or exceed the upper temperature limit of reactor operation. The coolant should, of course, be thermally stable at this temperature.

Heat Transfer Coefficient: The rate of heat flow from fuel to coolant is given by the expression $Q = hA \Delta t$ where Q is heat flux, h the heat transfer coefficient, A the area of contact and Δt the temperature difference. As this same relationship holds in the transfer of heat from coolant to working fluid, the advantage of a large h in a power reactor coolant is apparent. For a given heat flux and transfer area, a large h permits a smaller Δt and an increase in the overall conversion efficiency of the system.

Density and Specific Heat: The ability of a coolant to transport heat out of a reactor can be expressed by the equation $Q = \rho C_p A v \Delta t$ where Q is

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the heat absorbed, ρ is coolant density, C_p its specific heat, A is coolant cross sectional area, v is flow velocity and Δt is the temperature rise of the coolant. The product ρC_p is the heat capacity (or ability to store heat) of the coolant, and the larger this product the more effective will be the coolant as a heat recovering agent. For a given rate of heat removal, an increase in heat capacity will permit a decrease in flow rate and consequently a decrease in power expended for coolant circulation.

Neutron Capture Cross Section: Low neutron capture cross section in a coolant is desirable to enhance neutron economy and increase the specific power rating of the reactor. Good neutron economy minimizes the requirement for fuel enrichment and increases the burn-up time for fuel elements through increased production of fuel in fertile material and through the availability of spare neutrons to compensate for those lost in poisoning by fission fragments.

Moderating Power: A coolant with high moderating power may serve a dual purpose in a thermal reactor, for if its neutron capture cross section is sufficiently small, it may be used as moderator as well as coolant. Such material will lend itself to the design of inherently safe thermal reactors, inasmuch as the loss of coolant is accompanied by a reduction in the total moderating power of the reactor, and a consequent reduction in its reactivity. The ability of a coolant to moderate neutrons should reduce the amount of moderator otherwise required, and permit some reduction in reactor dimensions.

Radiation Stability: Radiation, other than neutrons, accompanying the fission reaction should not adversely change the physical properties or cause dissociation of a compound used as coolant. The presence in appreciable

quantity of undesirable radiation products in the coolant will necessitate measures for their removal, and add to the complexity of the cooling system.

Induced Radioactivity: When bombarded by neutrons, all elements except helium undergo transmutation to form isotopes of one greater mass number. Many of the isotopes so formed are radioactive, decaying with beta or gamma emissions or both to form a new element, or to the ground energy level of the isotope formed. Beta decay presents no problem, but the presence of hard gamma ray emitters in the transmuted coolant atoms will necessitate the addition of heavy shielding to the cooling system external to the reactor. Unless the half lives of these gamma emitters is very short, access to the cooling system after shutdown will be delayed or very restricted for a considerable period. A low level of activity and short half life in the neutron-activited coolant is obviously advantageous.

2. Gas as a coolant.

Because of their poor thermal qualities (i.e., low boiling point, low density, low specific heat and small heat transfer coefficient) gases are not generally satisfactory as power reactor coolants. Helium, which has a high specific heat, is a notable exception to this rule and can, by putting the cooling system under about ten atmospheres pressure, be made to yield 265 p.s.i.a. super heated steam at 525° F in a graphite-uranium reactor [14].

Helium has two advantages over other coolants; it is chemically inert and has a zero neutron capture cross section. Therefore, it is not subject to radiation damage or to induced radioactivity, and offers no corrosion problems. However, it is difficult to maintain helium in a large system under pressure, and the expenditure of power in circulating the gas reduces the efficiency of converting nuclear energy to useful power.

3. Water as a coolant.

Water has in fair measure all of the thermal properties of the ideal coolant save that of a high boiling point. It has a conveniently low melting point at 32° F, a reasonably large heat transfer coefficient, a high specific heat at the near constant value of 1.000, and is relatively dense compared to other liquids. The heat capacity (ρC_p) is larger than that of other available coolants, and a modest expenditure for pumping power will circulate it at adequate rates for removal of large quantities of heat. The low boiling point and high vapor pressures of water are its most serious thermal drawbacks as a coolant. Water boils at 212° F and one atmosphere pressure (14.7 p.s.i.a). To remain liquid at 650° F it must be kept under pressure in excess of 2200 p.s.i.a. and above its critical point at 706° F and 3226 p.s.i.a., water ceases to exist in the liquid state. It is obvious then that to attain reasonable thermal efficiencies with water as a power reactor coolant, relatively expensive high pressure systems will be required.

At high temperatures water is extremely corrosive of usual structural materials and, for reasons as will be subsequently explained, the use of corrosion inhibitors in the cooling water of a nuclear reactor is undesirable. All materials, even the highly corrosion resistant ones that must be used in the cooling system of a reactor, are to some extent attacked by water. Since a high degree of purity is required in the coolant for radiation stability, as well as to keep down the level of induced radioactivity, means must be provided to remove these corrosion products. Corrosion accellerating gases, particularly oxygen, must also be removed from water.

Many of the nuclear properties of water make it desirable for use in a thermal reactor. Because of its high neutron scattering cross section and

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large slowing power, water is an effective neutron moderator. However, its value as a moderator is diminished by the rather large thermal neutron capture cross section of hydrogen. In reactors utilizing enriched fuel, water can function effectively as both moderator and coolant, and it is significant to note that in the Submarine Thermal Reactor, which first demonstrated the practical application of nuclear power, water was used in this dual capacity.

In some reactor applications the radiation stability of water should be considered. Under the influence of high energy radiation water tends to decompose to hydrogen and oxygen gases, with the formation of small amounts of hydrogen peroxide. This effect of radiation on water has been investigated by Allen [1] , and found to be a function of the specific ionization of the radiation to which it is subjected and of the concentration of impurities which it holds in solution.

Intermediate to decomposition is the production of free radicals H and OH, which in pure water interact with each other and their product molecules in two competing reaction. One, the forward reaction, leads to the formation of hydrogen gas (H_2) and hydrogen peroxide (H_2O_2), which itself undergoes partial decomposition on further reaction with free radicals to form oxygen gas (O_2). The other, or back reaction, brings about the regeneration of water. The rate at which either of these reactions proceeds depends partly on the ionization density associated with a particular radiation. In the case of gamma or beta radiation, ionization density is low and free radicals are rather uniformly distributed. The chance is then good that unlike radicals will combine to regenerate water, so that for these radiations the back reaction is highly favored, and pure water appears to be

quite stable. On the other hand, more heavily ionizing particles such as protons, alpha particles or fission fragments cause local concentrations of like radicals which interact to form decomposition products. When pure water is subjected to these strongly ionizing radiations the back reaction is unable to compete with the forward reaction, and water decomposes with the evolution of hydrogen and oxygen gases.

Impurities dissolved in water are capable of reacting with free radicals. Their effect is to use up the free radicals which would enter the back reaction, and to accelerate decomposition. Particularly reactive in this regard are materials which are readily oxidized or reduced, such as corrosion inhibitors. Dissolved oxygen also retards the back reaction and, as it is one of the principal corrosive agents in water, its presence in reactor cooling water is doubly undesirable.

In a nuclear reactor other than the homogeneous type, water is primarily affected by only neutrons and gamma radiation. Neutrons themselves are not ionizing, but they produce ionization in water by the activation of protons on collision with hydrogen nuclei. In the ionization produced by protons the forward reaction is not as strongly favored as is the back reaction for gamma ionization. It appears unlikely then that pure water will undergo extensive decomposition in a heterogeneous reactor, but to maintain this stability water must be kept in as pure a state as possible.

The high neutron flux in a thermal reactor causes water to become intensely radioactive as a result of the (n, p) reaction with O^{16} to form N^{16}. The N^{16} decays with a half life of 7.5 seconds, and in the process emits hard gamma rays of 6.2 and 6.7 Mev. The short half life of this reduced radiation makes the cooling system accessible within a few minutes after
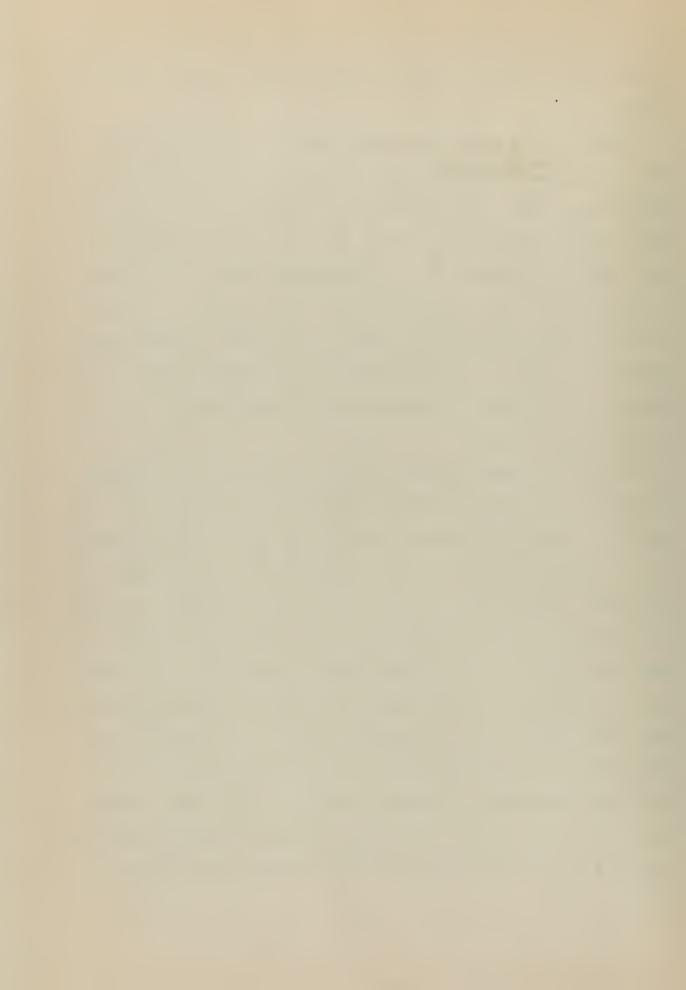
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shutdown, but during operation the entire cooling system external to the reactor must be heavily shielded to protect personnel from the high energy gamma.

Essentially the same problems exist with the use of heavy water as with light water. In regard to thermal properties, corrosion, radiation stability and induced radioactivity, the two fluids are nearly identical. The principal advantage of heavy water lies in its very low thermal neutron capture cross section and its high moderating power (it is the best moderator known). These properties enable it to be used as both moderator and coolant in reactors using natural uranium fuel. Its chief disadvantages are its high cost (about $80.00 per pound) and the special handling required to prevent its contamination with light water.

4. Water coolant systems.

It may be concluded that water can be used as coolant in a thermal neutron power reactor if low thermal efficiencies and saturated steam are acceptable in the power generating plant. To be used for this purpose the reactor coolant will have to operate in a closed, high pressure recirculating system. All welded construction of the most highly corrosion resistant materials is required to insure absolute leak tightness, and to minimize induced radioactivity. The cooling water must be maintained at the highest possible state of purity. This requires filtration and demineralization in the recirculating system, and deaeration as well as purification of the make up water, which should be stored under an inert gas blanket to prevent oxygen contamination. Provision should be made for system off-gassing as an emergency measure, in the event contamination of its cooling system should cause decomposition of water by the radiations to which it is exposed.

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The foregoing system requirements presume light water cooling of a heterogeneous reactor, moderated by water or heavy water. Such systems are inherently safe, or can be made so by the use of safety valves under the liquid moderator tank. In the water-cooled, water-moderated system loss of coolant would mean loss of moderator and reduction of reactivity. In the heavy water moderated, water cooled reactor the increased reactivity resulting from loss of water coolant would cause boiling of the heavy water moderator, and increased pressure in the core tank would activate the safety valve to dump moderator to a holding tank below the core. Experiments by Zinn at the Idaho Test Site $/12/$ have shown that no serious damage would result to such a reactor.

Attempt should not be made to utilize high pressure light water as coolant in a solid moderated reactor unless positive measures can be taken to prevent loss of pressure on the cooling system. Sudden loss of pressure could cause the coolant in the core to flash into steam, resulting in a nearly instantaneous increase in reactivity, and consequently a reactor casualty. This would not be true if heavy water were used as coolant, for loss of water would mean reduction in moderator (D_2O is the best moderator known), and reduction in reactivity.

Systems using heavy water as moderator and coolant could tolerate no loss of coolant, as the cost of make up heavy water is prohibitive.

Homogeneous reactor system may also use water or heavy water as combined moderator coolants. Such systems would require less elaborate purification equipment, but considerably more elaborate off-gas equipment. In addition, gas separation is required for removal of the gaseous radioactive fission products, and gas recombination is required to remove the explosive hydrogen (or deuterium) and oxygen gas mixture. The satisfactory operations

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of the HRE at the Oak Ridge National Laboratory has demonstrated the feasibility of such a system for power generation.

5. Liquid Metal coolants

The high temperatures available in a nuclear reactor have lead to a considerable amount of investigation into the application of certain low melting point, high boiling point metals and alloys to high temperature heat transfer systems. The developmental work thus far done has been largely confined to the alkali metals, lithium, sodium, potassium and an alloy of 56% sodium, 44% potassium by weight known as NaK. Some work has also been done on bismuth and the lead-bismuth alloy. However, only in the case of sodium and NaK has development reached the pilot plant stage /15/ . While much is yet to be learned about the technology of handling liquid metals, consideration should be given to the special properties of those mentioned above, which appear advantageous as power reactor coolants.

Two thermal properties of liquid metals, their high boiling points and their very large heat transfer coefficients, promise a significant increase in the efficiency of a nuclear power system. The high boiling point of the metals permits at least doubling of the maximum temperature of heat removal with water, and eliminates the necessity for high pressure plumbing. The larger value of h permits a smaller temperature difference between reactor and coolant for an equivalent quantity of heat transferred.

In regard to density and specific heat the metals suffer by comparison with water. Bismuth and the lead-bismuth alloy, which are approximately 13 times as dense as water at 500° F, have considerably lower heat capacities because of their very low specific heats. The alkali metals (except lithium), which are comparable in density to 500° F water, have specific heats from a fifth to a third that of water, and have correspondingly lower

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heat capacities. Lithium has a specific heat equal to that of water, but is only half as dense; it has the largest heat capacity of the liquid metals. For equivalent volumetric rates of flow the light metals compare favorably with water as to pumping power requirements, but because of their lower heat capacities, increased flow rates will be required to take advantage of their high temperature characteristics. The heavy metals do not compare as favorably in pumping power requirements.
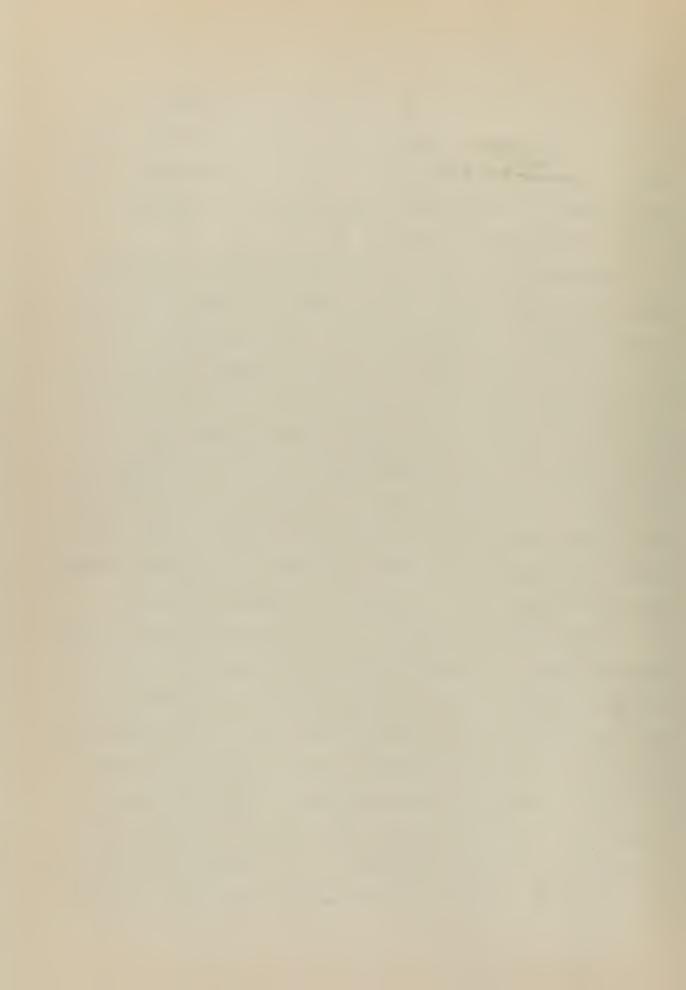
A unique feature in pumping liquid metals is the use of completely sealed electromagnetic pumps having no moving parts. These pumps work on the same principle as electric motors, i.e., force is produced on a conductor carrying current in an electric field. By sending a current through the liquid metal normal to its direction of flow and normal to a strong magnetic field, the metal is made to flow at acceptable rates.

The high melting point of liquid metals, most of which are solids at room temperature, requires the provision of preheaters in the cooling system. NaK, which melts at 12° F, is the notable exception to this rule. Mercury, which melts at minus 37° F, is not suitable as a coolant for thermal reactors because of its very large neutron capture cross section.

The alkali metals are among the most active chemical substances known. They burn readily in air and react violently with water, thus necessitating special precautions in handling at high temperatures and in preventing contact with the water used in steam generation. Bismuth and the lead-bismuth alloy require no special consideration in regard to reaction with water.

All of the liquid metals corrode the high temperature structural materials used to contain them and the rate of corrosion is accelerated by the presence of impurities, particularly oxygen. Experience has shown that the problem of corrosion can be readily overcome in the case of the alkali

metals. When these metals have been purified by filtration methods at ordinary handling temperatures and sealed in a thoroughly cleaned closed system, corrosion is reduced to insignificant levels.

Absolute leak tightness is required in the liquid metal systems to reduce the external hazard of fire and prevent oxygen contamination of the coolant. Completely welded circuits of austinitic stainless steel and nickle have given the desired degree of leak tightness in tests with the alkali metal systems. Inert gas blanketing of free surfaces is required as a further measure to prevent oxygen contamination.

All of the liquid metals have relatively poor nuclear properties in one respect or another. None are satisfactory as moderators, and all of the alkali metals except sodium have neutron capture cross sections greater than water. Special attention must, therefore, be given to safety features in the design of reactors utilizing these materials as coolants. Bismuth alone has a capture cross section (15 millibarns) sufficiently low to be tolerated in a reactor without seriously affecting neutron economy.

All of the liquid metals are stable to radiations encountered in a nuclear reactor. Lithium, lead and bismuth have the particular advantage of not becoming radioactive with the emission of hard gamma rays when subjected to neutron bombardment, and present no problem in the external shielding of the cooling system. Sodium and potassium, however, are strongly activated by neutrons. The activated sodium has a half life of 15 hours, and emits gammas with energies of 2.76 and 1.38 Mev; the potassium decays in 12.4 hours with the emission of 1.5 Mev gammas. In addition to the obvious requirement for shielding systems using these materials, the systems will not be accessible for protracted periods after shutdown.

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Requirements for shielding a sodium or NaK coolant loop external to the reactor can be reduced by the use of an intermediate heat exchanger and a secondary coolant loop between the reactor coolant and the steam generating plant. Such a system would, of course, reduce the overall efficiency of nuclear power conversion.

In spite of its high level of neutron induced activity, sodium appears to be the most acceptable of the liquid metals now available for use as a thermal neutron reactor coolant. Considerably more experience has been had with the use of sodium as a high temperature heat transfer fluid than with other liquid metals. Its corrosion problems are understood and have been satisfactorily solved. (This is not true of bismuth). Its melting point of 208° F permits handling at reasonably low temperatures, and its other thermal properties permit removal of large quantities of heat at reasonable flow rates and at temperatures that are high enough for good thermal efficiencies in a steam plant operating on superheated steam. Its neutron capture cross section (approximately 3/4 that of water) permits its use in a reactor using only slightly enriched fuels, and problems of reactor safety from coolant loss considerations can be overcome by having both the inlet and outlet of coolant enter the reactor above the core. Use of a sodium cooled graphite moderated reactor for power generation has been studied by Parkins of the North American Aviation Company [10] .

What the future holds for liquid metal coolants depends on the progress of current and continuing research into the properties of the various metals. One matter worth investigation is the possibility of obtaining lithium in the form of the pure isotope Li^7, which has an abundance of 92% in natural lithium. This isotope, which has a neutron capture cross section

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of 33 millibarns, would be superior to sodium in every respect, except melting point. It would in fact be the nearest approach to the ideal coolant of any material whose properties are now well known.

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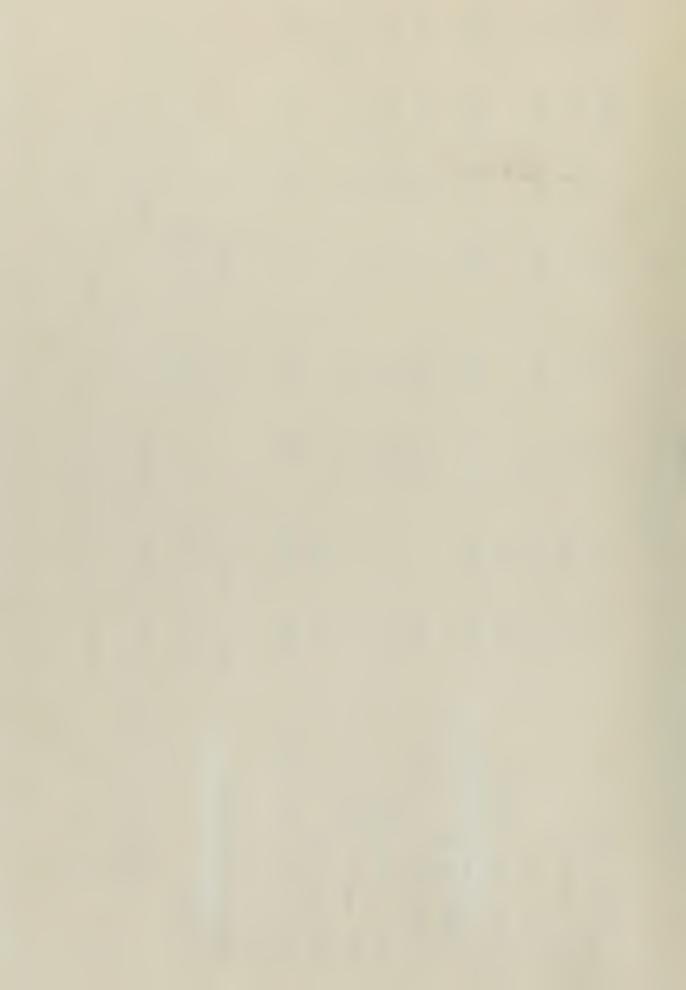
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APPENDIX I

PROPERTIES OF COOLANTS

Property\Coolant	H_2O**	D_2O	Li	Na	NaK	K	Pb-Bi	Bi
Melting Point °F	32	38.9	367	208	65	147	257	520
Boiling Point °F	212	214.7	2403	1616	1518	1400	3038	2691
Critical Temperature °F	706	701						
Heat transfer coefficient * $BTU/hr\ ft^2\ °F$	5970	5970	6330	9900	5820	7100	3950	4420
Density lbs/ft^3 at 500 °F	48.6		31.3	55.6	50.6	48.8	648	626
Specific heat $BTU/lb\ °F$	1.165		1.000	.320	.215	.187	.035	.035
Heat capacity BTU/ft^3	56.6		31.3	17.8	10.9	9.11	22.7	21.9
Moderating ratio	62	5000						
Capture cross section barns	.6016	.0009	67	0.45	1.1	2.5	0.17	.032
Induced radioactivity								
Quantum energies Mev	6.7-6.2	6.7-6.2		1.4,2.7	1.4-2.7	1.5		
Half life	7.5 sec	7.5 sec		15 hr.	12-15 hr.	12 hr.		

* From table 1. Reference 11--Based on coolant temperatures 525° F inlet 787° F outlet flowing in 1 2 inch diameter tube.

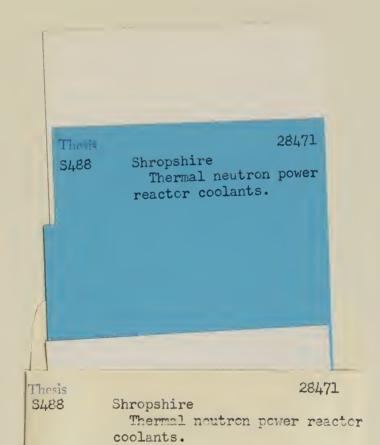** From tables 1 and 2. Reference 16. Remaining data primarily from reference 6.